Preparation of PAC Specimen by a Diffiusion Method

I. 13. Preparation of PAC Specimen by a Diffusion Method

Hanada R.

Institute for Materials Research, Tohoku University

Introduction

^{111}In nucleus is known to be the most convenient and appropriate for perturbed angular correlation (PAC) measurement. Several techniques have been applied to prepare the PAC specimen. Namely, (1) Implantation using a low energy accelerator. (2) Recoil implantation using nuclear reaction inside the specimen. (3) Recoil implantation using nuclear reaction at the surface of the specimen. (4) Diffusion.

The technique (1) is most frequently applied since any PAC specimen can be prepared without worrying about the solubility of In in the specimen. The technique (2) is applicable when a high energy accelerator as a cyclotron is available, although the specy of the metal specimen is limited. For instance only Cd or Ag can be used as the specimen when only a proton or α-particle beam is available. This technique has been applied by the present author for Cd metal. The technique (3) is applicable for any specimen in principle, although it is not efficient enough to prepare PAC specimens in a reasonable short time. Such an example for a Fe specimen is reported in a separate report. Also radiation damage always takes place in techniques (1)-(3), which may disturb the physical properties one wishes to study.

In the present the technique (4), namely, a diffusion method is applied to several kind of metal specimens of which results will be reported in the followings.

Experimentals

Metal specimens are purchased or prepared usually in a form of a thin foil 1 cm × 1 cm × 0.1 mm. Specimens are; group (4) Ti, Zr, Hf (5) Nb, Ta, V (6) Mo, W (8) Fe (9) Co (10) Ni, Pd, Pt (11) Cu, Ag, Au (13) Al (14) Si.

A few tenth of 1 ml of ^{111}In Cl aqueous solution is dropped on the surface of a specimen and dried at 373 K. After measuring the magnitude of the activity (A_0) at a constant distance, the specimen is annealed at 1100 K for 2 hrs usually in 1 atm of

hydrogen atmosphere. For the case of hydride forming specimens as Ti, Zr, Hf, Nb, Ta, V they are annealed in an ultra high vacuum. For the case of Al the annealing temperature is 700 K. After the annealing, the magnitude of the activity is measured again. (A_d) The ratio A_d/A_0 is used as a measure how much activity diffuses into the specimen. In some cases specimens are washed by water to examine the InCl solution is left on the surface. For the case of 1100 K annealing no trace of it is found.

Results and Discussion

Figure 1 shows the result of activity measurement where the ratio (A_d/A_0) for each metal specimen is plotted against the In maximum solid solubility in it. The solubility is obtained by referring a binary phase diagram.[1] Figure 1 shows a fair correlation is present between the ratio and the solid solubility of In. Namely the metal with a higher solubility shows a higher ratio. Among specimens investigated in the present, Pd has the highest ratio. When annealed with other specimens at the same time the ratio is found sometimes even to exceed 1 showing Pd attracts ^{111}In from others. The high ratio of the group (4) Ti, Zr, Hf, (10) Ni, Pd, Pt (11) Cu, Ag, Au indicates that ^{111}In diffuses into them. In order to examine this, PAC spectrum measurement is performed for each specimen. Figure 2 shows the results for Ti, Zr and Hf and Fig. 3 for Au.

Ti, Zr, Hf have a hexagnal close packed structure with the axial ratio different from the ideal one ($\sqrt{8/3}$) and hence a finite electric field gradient (EFG) is present at the substitutional site. This EFG causes a precession of ^{111}In nucleus to give rise to the rotation pattern as in Fig. 2. The presence of the rotation pattern in Fig. 2 reveals that ^{111}In diffuses into the bulk of them. The period of the pattern is though much longer than that of Cd showing the EFG in Ti, Zr, Hf is much smaller in magnitude than that of Cd.

Ni, Pd, Pt, Cu, Ag and Au have a cubic structure and hence no precession should take place with the null EFG. For the case of the ferromagnetic Ni a magnetic hyperfine field is present to cause the precession, of which result will be described in a separate report with that of Fe.

The result in Fig. 3 for Au shows an almost flat spectrum right after the specimen preparation (top) showing ^{111}In is in the site with no EFG. This is as expected for Au; cubic crystals. The specimen is annealed in air at several temperatures. The purpose of the annealing is to oxidize the specimen to find evidence of In-O complex formation. As is well known the restivity ratio of Au is increased when annealed in air and this has been ascribed to internal oxidation to cause an apparent purification of it. The spectra for Au annealed above 1073 K in Fig. 3 indicate an EFG distribution takes place in the specimen. Namely, a small hump takes place near delay time 0. This result indicates that oxygen atoms diffuse into the bulk above 1073 K to form In-O complexes with different number of

oxygen atoms thus giving rise to the EFG distribution. This shows that ^{111}In has diffused into the bulk during the diffusion annealing treatment. Further annealing in pure oxygen atmosphere at 973 K (bottom) causes some structures in the spectrum although the statistics are not high enough to conclude it as a real oxidation effect.

The present experiment shows that the PAC specimen can be prepared for Ti, Zr, Hf, Cu, Ag, Au, Ni, Pd and Pt by the diffusion method. Unfortunately, however, the method is found not applicable to Fe, Co, W, Mo, V, Nb, Ta, Al and Si in the present way of the treatment.

Reference

1) Binary Alloy Phase Diagram, ed. by Massalski T. B. (A.S.M. 1986).

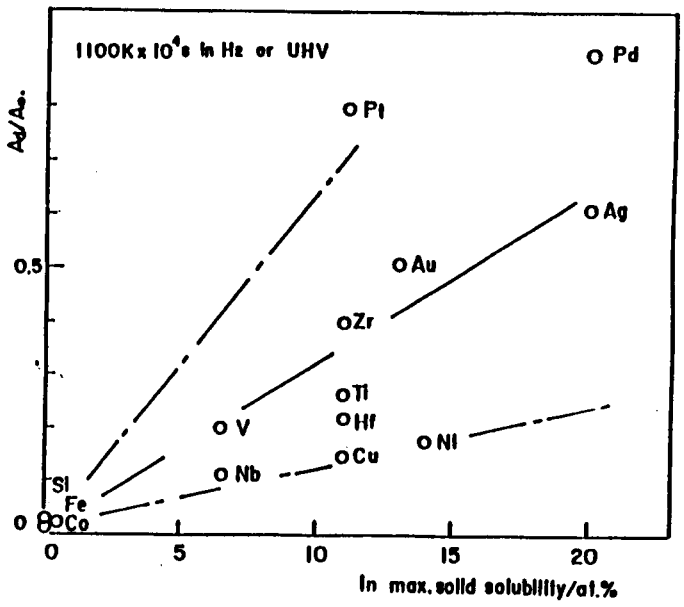

Fig. 1. The ratio Ad/Ao vs the maximum In solid solubility.
Ao: ^{111}In activity prior to the diffusion annealing
Ad: the same after the annealing.

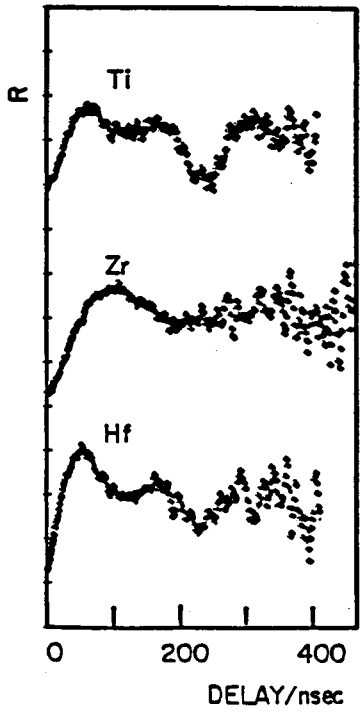

Fig. 2. PAC spectra $(N(\pi)-N(\pi/2))/(N(\pi)+N(\pi/2))$ for Ti, Zr and Hf.

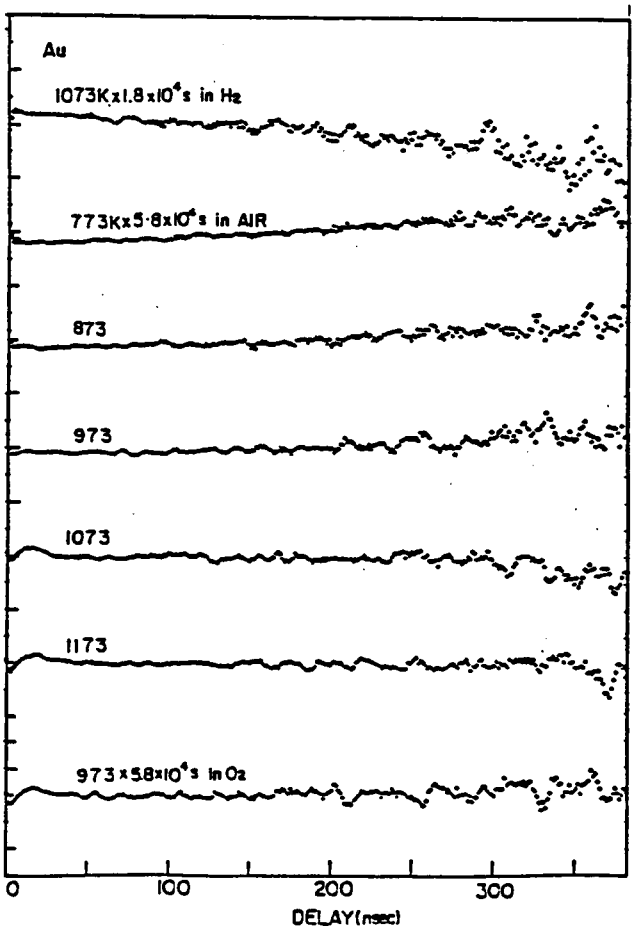

Fig. 3. PAC spectra for ^{111}In doped Au.